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TITLE: PASSIVE NEUTRON MEASUREMENTS AND CALCULATIONS OF IRRADIATED PWR FUEL ASSEMBLIES

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PASSIVE NEUTRON MEASUREMENTS AND CALCULATIONS OF IRRADIATED PWR FUEL ASSEMBLIES

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ABSTRACT

Passive neutron measurement of spent-fuel assemblies is a convenient and rapid technique for verifying operator-declared exposure values. Experimental results have been obtained for a wide variety of pressurized-water reactor fuel assemblies at reactor and away-from-reactor storage facilities. Calculations using a single-point depletion code have been performed to evaluate and quantify the effects of various initial fuel parameters and irradiation histories on the neutron source strengths in spent-fuel assemblies.

1. Introduction

Passive neutron measurement of spent-fuel assemblies is a convenient and rapid technique for verifying operator-declared exposure values to satisfy both safeguards and facility-operator requirements. The instrumentation is simple to operate and the acquired data are reasonably easy to analyze. To demonstrate the applicability of the technique to satisfy International Atomic Energy Agency (IAEA) safeguards requirements and to establish a data base, ongoing measurements are being made on various spent-fuel assemblies. To date, 36 pressurized-water reactor (PWR) fuel assemblies have been examined at a reactor storage facility, and 17 assemblies from four different reactors have been examined at an away-from-reactor (AFR) facility.

Parametric studies were performed to quantify the effects of initial fuel parameters and irradiation histories on the depletion and production of actinide isotopes in spent-fuel assemblies. A calculational model based upon a PWR-type design was used for these investigations.

2. Experimental Measurements

Power Reactor Storage Facility.

Neutron emission rates for 36 PWR fuel assemblies with declared exposures ranging from 18.8 to 38.0 GWd/tU were measured.[1] The detector consisted of two fission chambers (165 mg ^{235}U each) placed horizontally on opposite sides of the fuel assembly to reduce positioning effects. Cooling times since discharge ranged from 4 to 40 months. Initial ^{235}U enrichments were 2.25, 2.80, and 3.30 wt% for exposures at 20, 30, and above 35 GWd/tU, respectively. Results from these measurements are shown in Fig. 1 in which the 4-month data have been denoted differently. There are two principal sources of neutrons in spent-fuel assemblies above 10 GWd/tU; namely ^{242}Cm ($t_{1/2}$ = 0.4456 yr) and ^{244}Cm ($t_{1/2}$ = 18.1 yr). Because of the half-life differences of these two isotopes, neutron emissions for assemblies with long cooling times (>2 yr) are primarily from ^{244}Cm. For short cooling times both isotopes contribute. The influence of ^{242}Cm is shown in the 4-month data in Fig. 1 by increasing the measured neutron rate above that which would be expected if only ^{244}Cm were contributing to the rate.

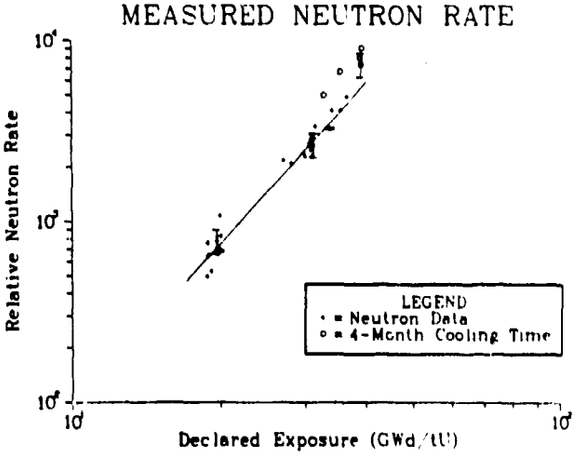

Fig. 1. Experimental results for 36 PWR spent-fuel assemblies having cooling times of 4 to 40 months.

For exposures above 10 GWd/tU, data like those in Fig. 1 can be represented by a power-function correlation of the form:

$$\text{Neutron emission rate} = a\,(\text{exposure})^{\beta}, \quad (1)$$

where a is a scaling parameter and β ranges in value from 3.0 to 4.0 depending on the fuel being measured. Using this relationship the slope of the line given in Fig. 1 is 3.0.

Away-From-Reactor Storage Facility

A spent-fuel measurement system was installed in the receiving pool of an AFR storage facility to demonstrate the applicability of passive neutron techniques in a facility that had a very limited amount of available space. The system (Fig. 2) consisted of a pipe containing the detectors and a V-shaped positioning device into which the corner of the fuel assembly could be correctly positioned. The detector tube (Fig. 3) consisted of three detectors: (1) a fission chamber to measure the neutron emission rate, (2) an ion chamber to determine the gross gamma dose rate, and (3) a Be(γ,n) detector that is sensitive only to gamma rays with energies above 1.66 MeV. This detector package was used to measure the neutron emission rates of 17 spent-fuel assemblies from 3 pressurized-water and 1 boiling water reactors. The exposure range was 10-40 GWd/tU for the PWR assemblies and 5 GWd/tU for the boiling-water reactor (BWR) assembly. Two objectives of

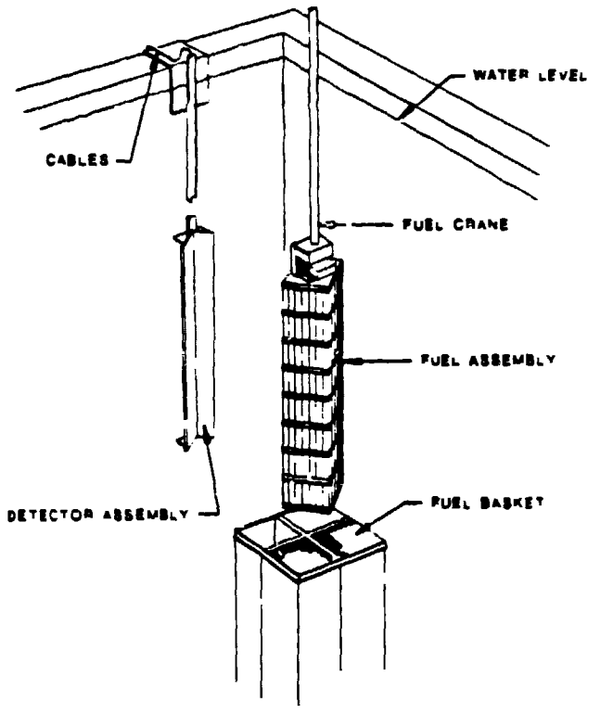

Fig. 2. Positioning device and detector tube assembly installed at the AFR facility to measure spent-fuel assemblies.

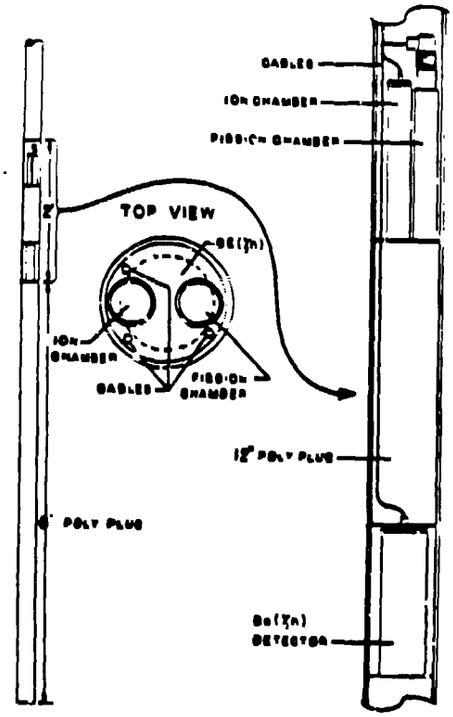

Fig. 3. Schematic of the detector package used to measure PWR and BWR fuel assemblies at the AFR facility.

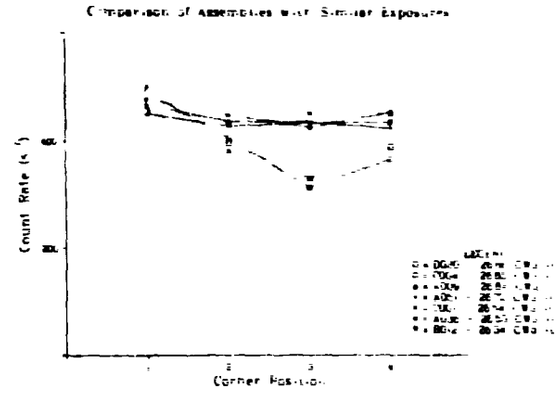

Fig. 4. Relative neutron count rates of the four corners of seven PWR fuel assemblies.

the measurements were the determination of the reproducibility of replicate measurements and the quantification of differences in measurements obtained at the four corners of the same fuel assembly. Precisions of 1.0% for the neutron, 0.5% for the gross gamma, and 1.5% for the $Be(\gamma,n)$ measurements were obtained.

Variations in the neutron measurements obtained at the four corners at the same axial position are shown in Fig. 4 for a set of seven PWR fuel assemblies with similiar exposures. While differences in the individual corner measurements varied up to 40%, the sum of the four corners varied from assembly to assembly by no more than 15%.

3. Calculational Results

Neutron Sources in PWR Fuels.

The production and depletion of actinide isotopes have been calculated for typical PWR-type fuel assemblies using a single-point depletion code.[2,3] Exposures from 0 to 50 GWd/tU were simulated to represent the complete range of exposures that an experimenter may have to measure. Atom densities for each actinide isotope were calculated at 35 exposure steps.

Table I shows a comparison of the calculated and experimentally measured values[4] for a selected set of isotopes. The agreement between the two values is very good, with the isotopes having large errors being more difficult to measure experimentally.

Neutron source rates from each isotope were calculated as a function of exposure and cooling time (up to 120 months). Representative results from these calculations are given in Figs. 5 and 6 for exposures of 10.8 and 29.5 GWd/tU. As mentioned in the previous section, the two curium isotopes, ^{242}Cm and ^{244}Cm, are the dominant sources of neutrons for fuel assemblies with exposures above 10 GWd/tU. Because of the 0.4456-yr half-life of ^{242}Cm this isotope is only a significant contributor during the first two years after discharge from the reactor. Therefore, ^{244}Cm is the principal source of neutrons in most light-water spent-fuel assemblies. The dominant influence of ^{244}Cm is seen in Fig. 7 as the curves merge after two years of cooling

TABLE I

COMPARISON OF MEASURED AND CALCULATED VALUES

Quantity	Measured[4] Value	Calculated Value	Percent Difference*
Burnup			
At.% Fission	3.221	3.221	+0.00
Exposure GWd/tU	30.92	31.19	+0.89
U-234/U	0.00014	0.00012	-12.81
U-235/U	0.00612	0.00588	-3.84
U-236/U	0.00352	0.00357	+1.52
U-238/U	0.99022	0.99043	+0.02
Pu-238/Pu	0.01676	0.01452	-13.35
Pu-239/Pu	0.54261	0.53966	-0.54
Pu-240/Pu	0.25101	0.24002	-4.38
Pu-241/Pu	0.12998	0.13772	+5.95
Pu-242/Pu	0.05964	0.06807	+11.41
Pu-239/U-238	0.00518	0.00496	-4.28
Nd-148/U-238	0.00057	0.00059	+3.96

*Isotopes with large errors (>10%) are difficult to measure experimentally.

time. The slope of a power function (Eq. 1) fitted to the calculational results between 10 and 50 GWd/tU is 3.6. These neutron source rates do not include any correction for transport of the neutrons through the fuel material and water to the detector.

Calculationally estimated values for ^{244}Cm correlate very well with the total fissile inventory in the fuel material as shown in Fig. 8. Figure 9 shows a similar plot of the total source neutron rate as a function of the remaining fissile inventory in a fuel assembly for various cooling times. The functional relationship is single-valued and linear between 10 and 50 GWd/tU on the semi-log plot. Similar plots of neutron source rate vs total plutonium and vs total plutonium minus ^{239}Pu are shown in Figs. 10 and 11, respectively. In each of the plots (Figs. 9-11) the influence of ^{242}Cm on the neutron

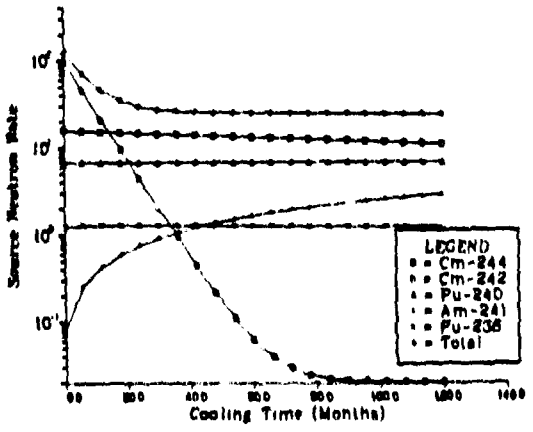

Fig. 5. Neutron source rate for the five most prominent contributors to the total rate for an exposure of 10.8 GWd/tU.

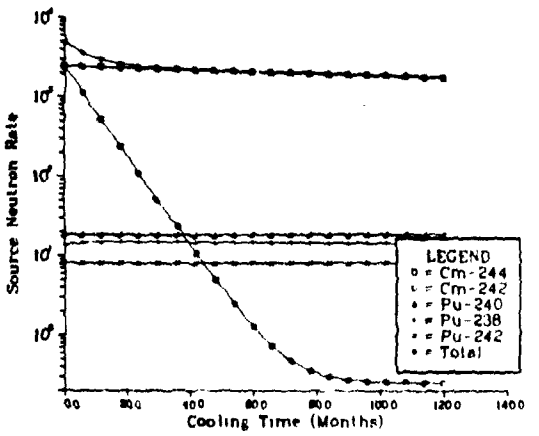

Fig. 6. Neutron source rate for the five most prominent contributors to the total rate for an exposure of 29.5 GWd/tU.

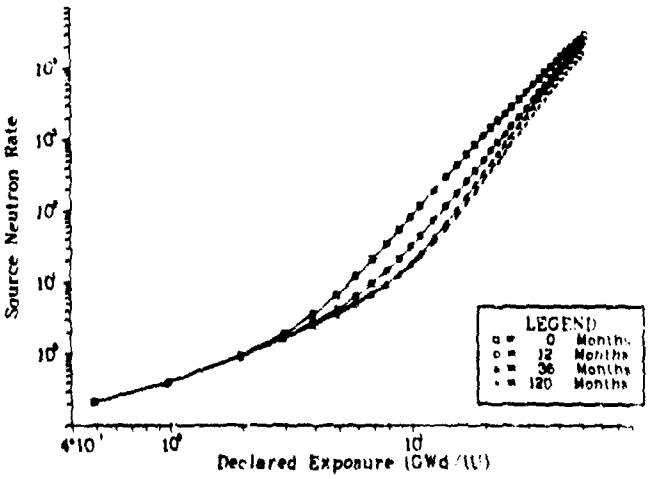

Fig. 7. Neutron source rate as a function of exposure for various cooling times.

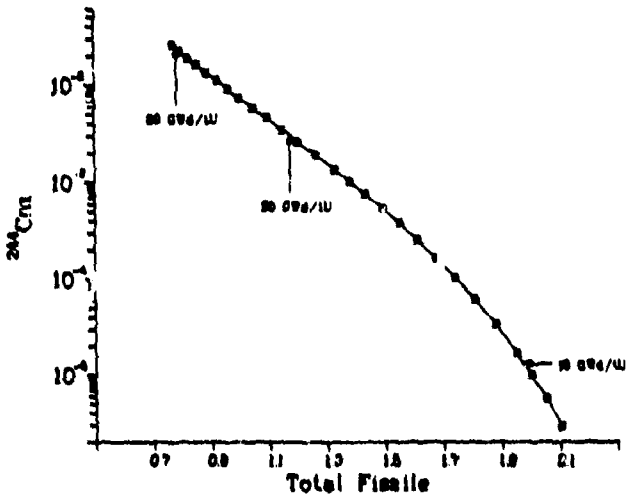

Fig. 8. Correlation of the ^{244}Cm isotopic concentration with the remaining fissile inventory in the fuel assembly.

source strength is only significant for short cooling times (<2 yr). Such correlations have been used to successfully predict plutonium content in spent-fuel assemblies.[5]

The isotopic composition of the plutonium changes significantly as a function of exposure. Figure 12 shows the percentage of total plutonium for each isotope as exposure increases. At 50 GWd/tU, ^{240}Pu would be expected to be about one-half as large as the ^{239}Pu fraction. With curves such as those in Fig. 12 and information on total plutonium content (obtained from ^{244}Cm vs total plutonium correlations), estimates could be made on plutonium isotopic composition.

4. Conclusions

Applicability of passive neutron measurements of spent-fuel assemblies has been evaluated experimentally and calculationally. Experimental results for 36 PWR fuel assemblies indicate that the measured neutron rate correlates with operator-declared values of exposure as a power function. Precise measurements can be obtained rapidly, requiring only 1-2 minutes measurement time. These measurements must be performed on more than one corner or side of the fuel assembly because significant differences have been found between corners of the same fuel assembly.

Calculational techniques have been used to examine correlations between the ^{244}Cm concentration and the total fissile material and total plutonium remaining in the fuel assembly. These calculational techniques have allowed us to explore the applicability of these correlations and other correlations in an extended exposure range (50 GWd/tU) where experimental data are not presently available.

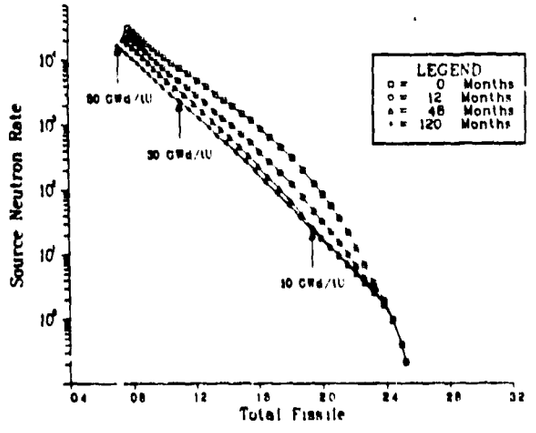

Fig. 9. Total neutron source rate as a function of the weight per cent of the total fissile material remaining in the fuel assembly for four cooling times.

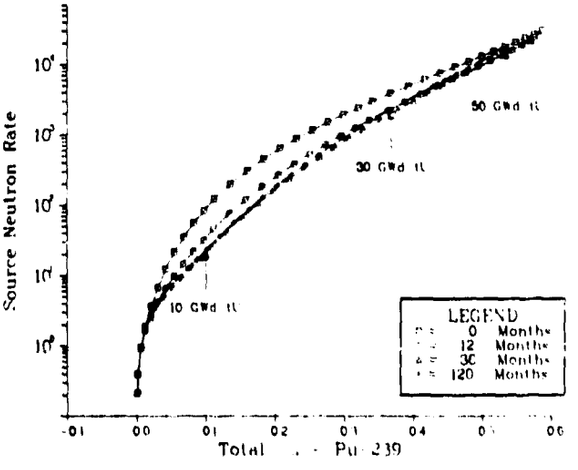

Fig. 11. Total neutron source rate as a function of total plutonium minus ^{239}Pu remaining in the fuel assembly for four cooling times.

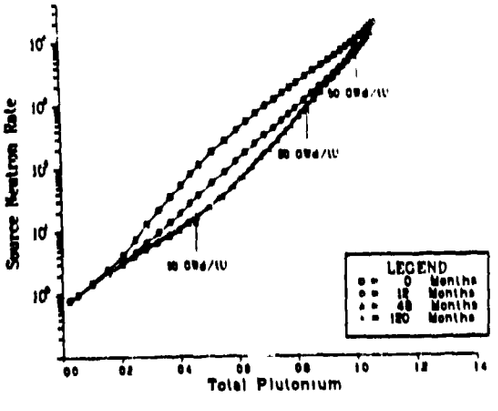

Fig. 10. Total neutron source rate as a function of the weight per cent of the total plutonium remaining in the fuel assembly for four cooling times.

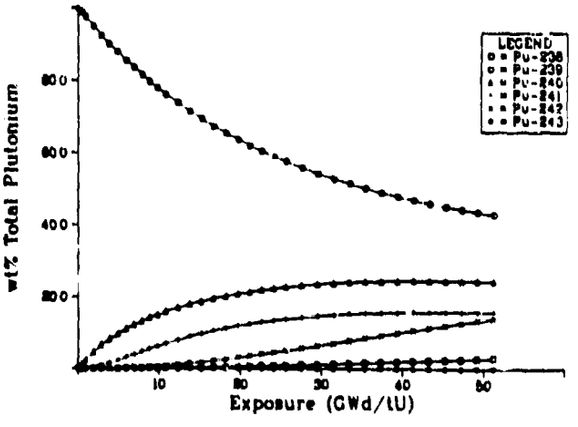

Fig. 12. Contribution of plutonium isotopes to the total as a function of exposure.

5. References

1. J. R. Phillips, J. K. Halbig, D. M. Lee, S. E. Beach, T. R. Bement, E. Dermendjiev, C. R. Hatcher, K. Kaieda, and E. G. Medina, "Application of Nondestructive Gamma-Ray and Neutron Techniques for the Safeguarding of Irradiated Fuel Materials," Los Alamos National Laboratory report LA-8212 (ISPO-77) (May 1980).

2. T. R. England, W. B. Wilson, and M. G. Stamatelatos, "Fission Product Data for Thermal Reactors, Part 2: Users' Manual for EPRI-CINDER Code and Data," Electric Power Research Institute report EPRI NP-356, Part 2 (Dec. 1976). Also published as Los Alamos National Laboratory report LA-6746-MS (Dec. 1976).

3. T. R. England, W. B. Wilson, and M. G. Stamatelatos, "Fission Product Data for Thermal Reactors, Part 1: A Data Set for EPRI-CINDER Using ENDF/B-IV," Electric Power Research Institute report EPRI NP-356, Part 1 (Dec. 1976). Also published as Los Alamos National Laboratory report LA-6745-MS (Dec. 1976).

4. A. A. Bauer, L. M. Lowry, J. S. Perrin, "Progress on Evaluating Strengths and Ductility of Irradiated Zircaloy During July Through September, 1975," Battelle Memorial Institute report BMI-1938, p. 16 (1975).

5. G. Schulze, H. Wuerz, L. Koch, and R. Wellum, "Neutron Array Plus Isotopic Correlations: A Method for Determining Pu and Burnup in Spent LWR Fuel Assemblies," Proceedings of the 2nd Annual ESARDA Symposium on Safeguards and Nuclear Materials Management, Edinburgh, Scotland (1980), p. 396.